

# Turing structures in dc gas discharges

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Self-organized spatial plasma structures exhibit many similarities with Turing structures obtained in biology and chemistry. Using an analytical mesoscopic approach it is shown that plasma balls of fire belong to the same class of Turing structures like the Brusselator. It is also mathematically proved that the existence of these self-organized plasma structures is related with a negative differential resistance. There are also established the mathematical conditions which the negative differential resistance must satisfy for obtaining a stationary ball of fire at the anode of a plasma diode.

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## I. INTRODUCTION

In a paper published in 1952 [1], the British mathematician Alan Mathison Turing suggested a possible connection between biologic structures emerged during the morphogenesis process and chemical structures spontaneously formed in reaction-diffusion (RD) systems. For 15 years that paper did not have any echo among scientists, but starting with 1960s Ilya Prigogine's School from Brussels, Belgium begun an intensive study of Turing's idea [2]. The result was a theoretical chemical model, known today by the name of Brusselator [3]. In spite of these positive theoretical results, the first experimental evidences of chemical Turing structures were obtained only in 1990-1991 by the groups of Patrick de Kepper in Bordeaux, France [4] and of Qi Ouyang in Austin, Texas, USA [5].

The theoretical approach of Turing structures involves the study of the RD equations describing the system under study. The general method of treating this kind of mathematical problem is outlined in [6], [7] and it implies the use of the tools of nonlinear dynamics. Another mathematical and conceptual method for studying the Turing structures in solid state, but also in plasma physics, is by considering them as autosolitons [8]. Recently, with the help of numerical simulations, there was obtained a large variety of spatial patterns and structures in two and three dimensions, from spots and stripes, to lamellae and spherical droplets [9].

Almost 45 years after the publication of Turing's paper, a group of physicists from Münster University, Germany [10, 11] have experimentally observed that, as a result of dc discharges in a quasi-bidimensional geometry, self-organized luminous structures have been formed, in the form of spots, striated filaments, hexagons and stripes. The self-assembling of such patterns has been attributed to a Turing-type scenario, analogous to that encountered in chemical systems [8, 10, 11].

Developing some similar experiments with the above ones, in a different experimental arrangement, but also in a bidimensional geometry, we analyzed the succession of the physical processes at the basis of the current filamentation in plasma systems [12]. The experiments proved

that at the origin of current filaments is the formation of a quasi-bidimensional anode spot. Its emergence follows the same physical scenario as in the three-dimensional case, where it is also known as "plasma ball of fire" [13]. In the attempt to search if plasma balls of fire are Turing-type structures, our previous theoretical studies proved that taking into account elementary quantum processes, like excitation and ionization, which are the key ingredients for ball of fire formation, Turing-like structures cannot arise in plasma [14]. In other words, the above study proved once more that for describing the emergence of a self-organized structure the mesoscopic approach is necessary, the microscopic one being inadequate.

The aim of this paper is to prove analytically, on the basis of a mesoscopic approach (*i.e.* starting from the equivalent electric circuit of the plasma system), that the appearance of a ball of fire is associated with a negative differential resistance. The present results also prove, for the first time to the knowledge of the author, that the plasma ball of fire belongs to the same class of Turing structures like the Brusselator: activator - substrate depleted. There are also established the mathematical conditions the negative differential resistance must satisfy for obtaining a stationary ball of fire at the anode of a plasma diode.

## II. WHAT ARE TURING STRUCTURES?

In his seminal paper [1], Turing showed that, in certain conditions, a homogeneous biologic medium can undergo a spatial symmetry breaking, giving birth to a spatial ordered structure. The present understanding of the concept of Turing structure is mainly due to de Kepper's group [4]: Turing structures are self-organized stationary spatial structures appearing in dissipative systems and correspond to the stationary stable solutions of the RD equations modelling the studied physical system. Turing structures appear only in open systems [1], far from the thermodynamic equilibrium, where the essential processes - reaction and diffusion - can be coupled. As already proved [6], one of the key processes for the appearance of spatial structures is the autocatalytic one, *i.e.* the

self-enhancement of a chemical species, called *activator*. The other important species in the appearance of Turing structures is called *inhibitor*, or, in different circumstances, *substrate depleted*. Both, the activator and the inhibitor (or substrate depleted), are referred to as *intermediate species*. Finally, the rest of chemicals existent in the system are called *pool species* [7]. The destabilization of the stationary homogeneous state and the emergence of a Turing structure can be realized only if the diffusivities of the intermediate species are different [15]. If the stationary homogeneous state of an RD system is stable to small spatial perturbations in the absence of diffusion, but unstable when the diffusion process is present [6], then an instability will take place in the system. The result of this instability, now called Turing instability, is the spatial structuralization of the system. Compensating the difference between the reaction rates, the diffusion makes the inhomogeneous state be stationary [2]. Of course, this is valid as long as the system feeding is continuous, otherwise the structures are transitory.

More recently, it was demonstrated that another necessary condition for the appearance of Turing structures is the presence of cross-inhibition [16], that is an intermediate species inhibits the increase of another intermediate species' concentration.

### III. PLASMA BALLS OF FIRE

When a dc power supply is connected, through a load resistor, to a plasma diode, the processes taking place in front of the anode are numerous, they depending on the magnitude of the potential drop on the diode. The experiments clearly show that, as the plasma system is gradually departed from the thermodynamic equilibrium, the processes inside the diode become strongly nonlinear.

Gradually increasing from zero the potential drop on the plasma diode, the plasma system first behaves like an ohmic conductor, after that the static  $I(V)$  characteristic becoming nonlinear [17, 18]. The nonlinearity of the static  $I(V)$  characteristic is related with the quantum processes of excitation and ionization taking place in front of the anode. These processes give birth, in order, to an electron layer and a positive ion layer, the later being localized between the electronic one and the anode. The interactions between these layers are collective ones, the electrostatic forces acting between large groups of electric opposite charges. They lead to the self-assembling of a planar electric double layer in front of the anode. Increasing further the potential drop on the plasma diode, there will be a threshold potential drop for which the planar double layer transits into a spherical one for minimizing the free energy of the system. The spherical double layer, having the negative sheath at the exterior, covers a nucleus consisting of a plasma enriched in positive ions [Fig. 1(a)]. This luminous, beautifully colored, complex space charge configuration, formed at the anode surface, is known by the name of anode spot or plasma ball of fire

[17]. During the self-assembling of the ball of fire, which is a spontaneous process, the static  $I(V)$  characteristic displays a sudden jump, the current increasing abruptly, while the potential drop on the diode is at its threshold value.

Increasing the potential drop even slightly above the threshold value, the ball of fire exists in a stationary state at the anode surface. Decreasing now the potential drop on the diode, the ball of fire will exist in the stationary state even for potential drops smaller than the threshold value. This means nothing else than the existence of the ball of fire is displayed in the static  $I(V)$  characteristic by a hysteresis loop, which is one of the fingerprints of self-organization, proving that the structure has memory. The stable branches of the hysteresis loop coexist, meaning that the characteristic displays a region of bistability. Between these two stable branches there is another one, unstable, on which the differential resistance of the gaseous conductor is negative. Because the succession of these three states has the form of the letter S, the negative differential resistance is commonly called S-type negative differential resistance. This negative differential resistance proves that the ball of fire acts as an energy reservoir, allowing the structure to exist for a while, even for worse external conditions than those needed for its emergence.

### IV. CIRCUIT THEORY FOR A PLASMA DIODE WITH A BALL OF FIRE AT THE ANODE SURFACE

The experiments studying the emergence of the plasma ball of fire revealed the following: the double layer bordering the self-organized structure has an electric capacity [19]; the ball of fire has an inductance related with the inertial properties of the positive ions from its nucleus; last but not least, the ball of fire is responsible for the hysteresis loop appearance, as well as for electric oscillations in the anode circuit, which means that it acts as a negative differential resistance [17].

Based on the above experimental results and choosing the experimental arrangement such that only the anode part of the discharge to be important, the simplest equivalent electric circuit for the plasma diode can be established and it looks like that in Fig. 1(b). Because the ball of fire is a nonlinear circuit element, the potential drop on its dynamic resistance is given by a nonlinear function of the current, labeled by  $f(i)$  in Fig. 1(b). This function gives the S-form of the static  $I(V)$  characteristic of the plasma diode.

Under these circumstances, from the circuit equations  $[U = i_1 r + u; i_1 = i + C \frac{du}{dt};$

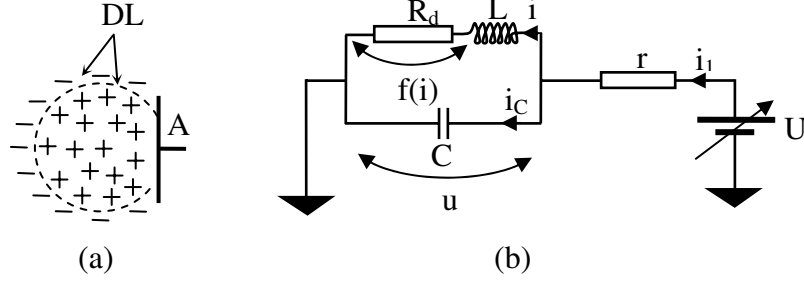


FIG. 1: (a) Schematic representation of plasma ball of fire. A - anode (disc electrode 3 cm in diameter), DL - electric double layer, plasma parameters: Ar, pressure 10 mTorr, density  $10^9 \text{ cm}^{-3}$  [18]; (b) Electric circuit of a plasma diode with a ball of fire on the anode.  $U$  is the variable voltage of the dc power supply,  $r$  is the load resistor,  $R_d$  is the dynamic resistance of the ball of fire,  $L$  its inductance,  $C$  its capacitance, and  $u$  the potential drop on the diode.

$u = L \frac{di}{dt} + f(i)$ , by eliminating  $i_1$ , we get

$$\begin{cases} \frac{di}{dt} = -\frac{1}{L}f(i) + \frac{1}{L}u \equiv F(i, u), \\ \frac{du}{dt} = \frac{U}{rC} - \frac{1}{C}i - \frac{1}{rC}u \equiv G(i, u). \end{cases} \quad (1)$$

The *rhs* in eqs. (1) give just the reaction functions  $F$  and  $G$  for the plasma system analyzed here. Adding the appropriate diffusion terms, the RD (or drift-diffusion) system of equations reads:

$$\begin{cases} \frac{\partial i}{\partial t} = F(i, u) + D_i \nabla^2 i, \\ \frac{\partial u}{\partial t} = G(i, u) + D_u \nabla^2 u. \end{cases} \quad (2)$$

Here  $D_i$  accounts for the diffusion of the positive ions and  $D_u$  for that of electrons [17].

In the stationary homogeneous state the solutions of the system are the fixed points [ $F(i, u) = 0$  and  $G(i, u) = 0$ ] given by

$$f(i_0) = u_0 = U - ri_0. \quad (3)$$

Since the stationary homogeneous state of the plasma system (*i.e.* when the ball of fire is not formed yet on the anode surface, but the system is on the lower branch of the bistability region) is stable, this agrees with the first condition required for the emergence of Turing structures: the stationary homogeneous state is stable to small homogeneous perturbations (*i.e.* in the absence of diffusion). By imposing this, we can find the conditions that must be satisfied by the system's equations when small perturbations act upon it (Lyapunov method). Let the perturbed values of  $i_0$  and  $u_0$  be  $i = i_0 + x$ ,  $x \ll i_0$  and  $u = u_0 + y$ ,  $y \ll u_0$ . As long as the fixed points of the system of equations are not hyperbolic (*i.e.* they are not bifurcation points), Hartman-Grobman theorem from Nonlinear Dynamics [20] is valid: "In an open neighborhood around a nonhyperbolic fixed point, the dynamics of the nonlinear system is topologically equivalent with the dynamics of the linearized

system". So, linearizing the static  $I(V)$  characteristic of the system around the fixed point  $(i_0, u_0)$ , we get:

$$f(i) = f(i_0) + \left. \frac{df}{di} \right|_{i=i_0} \cdot (i - i_0) + \dots \cong f(i_0) + R_d \cdot x,$$

where  $\left. \frac{df}{di} \right|_{i=i_0} = R_d$  is the dynamic or differential resistance of the nonlinear circuit element in the working point  $(i_0, u_0)$ .

In this way eq. (1) can be written

$$\begin{cases} \frac{dx}{dt} = -\frac{R_d}{L}x + \frac{1}{L}y, \\ \frac{dy}{dt} = -\frac{1}{C}x - \frac{1}{rC}y. \end{cases} \quad (4)$$

One necessary condition for the emergence of Turing structures is the presence of cross-inhibition [16]. Analyzing eq. (4), the cross-inhibition is present if the products of the diagonal elements of the Jacobian matrix of the above system are negative [16]:

$$\begin{cases} \left( -\frac{R_d}{L} \right) \cdot \left( -\frac{1}{rC} \right) < 0, \\ \left( \frac{1}{L} \right) \cdot \left( -\frac{1}{C} \right) < 0. \end{cases} \quad (5)$$

If the second inequality is evident, the first one is satisfied if and only if the differential resistance of the gaseous conductor is negative

$$R_d < 0. \quad (6)$$

Indeed, this result confirms the experimental finding according to which the ball of fire acts as a negative differential resistance in the diode's circuit. It must be stressed here that, although the above analysis is made for the stationary homogeneous state (*i.e.* when the ball of fire is not yet formed at the anode surface), the system is in the bistability region, where the third, unstable state also exists and it displays a negative differential resistance. Also, if the system is on the upper branch of the bistability region, for the same value of the potential drop on the plasma diode (*i.e.* the control parameter of the

system) as above, the ball of fire will exist at the surface of the anode.

Taking into account the above analysis, the matrix of signs associated with the Jacobian matrix of the system, evaluated in the stationary homogeneous state, reads

$$\text{sgn}J_0 = \begin{pmatrix} + & + \\ - & - \end{pmatrix}.$$

This corresponds to a model of Turing structure called activator - substrate depleted [21] and is of the same type as the Brusselator [7]. In the frame of this model the substrate is depleted during the autocatalysis process. Its consumption slows down the auto-amplification process of the activator concentration. Activator - substrate depleted systems are characterized by opposite signs for the couplings of a species with itself and with the other one. The result is a spatial distribution of concentrations in opposition of phase: in the place where one species concentration is maximal, the others concentration is minimal and vice versa. The concentration peaks for this type of system are not sharp, but rounded, the new peaks being formed by the splitting of the existing ones and their shifting [7]. The explanation is as follows: the substrate concentration is larger in the vicinity of the place where the activator peak grows by depleting the substrate. This determines the activator concentration to grow in lateral direction. As a result, the concentration peak of the activator will split up and shift towards the regions that have a higher concentration of substrate. This scenario explains the formation of several balls of fire on the anode [22, 23] as well as their mutual disposition on the anode, in the form of regulate polygons [24].

The stability of eqs. (2) to small homogeneous perturbations is ensured if the trace and the determinant of the Jacobian matrix  $J_0$  of eqs. (4), evaluated in the stationary state, satisfies concomitantly the following conditions [6]:

$$\begin{cases} \text{Tr}J_0 < 0, \\ \text{Det}J_0 > 0, \end{cases} \quad (7)$$

or

$$\begin{cases} |R_d| < \frac{L}{rC} = \frac{Z_0^2}{r}, \\ |R_d| < r, \end{cases} \quad (8)$$

where  $Z_0^2 = L/C$  is the proper impedance of the ball of fire. In conclusion, one necessary condition for the appearance of Turing structures is:

$$|R_d| < \min \left\{ r; \frac{Z_0^2}{r} \right\}. \quad (9)$$

Since  $Z_0$  always lies between  $r$  and  $Z_0^2/r$ , this means that the stationary ball of fire permanently self-adjusts its structure such that its negative differential resistance to be smaller than its proper impedance. The fact that the negative differential resistance of the ball of fire is

smaller than  $Z_0^2/r$  means that any small perturbation acting on the system extinguishes, the system behaving as a damped oscillator. The equation for this oscillator can be easily obtained by eliminating  $u$  from the second eq. (1) with the help of the last circuit eq. and the damping coefficient of this oscillator is  $\delta = \frac{1}{2L} \left( \frac{Z_0^2}{r} - |R_d| \right)$ . The perturbations' damping is ensured as long as the potential drop on the plasma diode does not surpass another critical value, at which the ball of fire disrupts [17]. Above this new critical value, the current variations become periodic (*i.e.*  $|R_d| = Z_0^2/r$  or  $\delta = 0$ ) and the double layer bordering the ball of fire periodically detaches from its surface and travels a certain distance towards the cathode. This new critical value of the potential drop on the plasma diode delimitates the border between spatial and spatiotemporal self-organization.

The second necessary condition for the appearance of Turing structures requires that the stationary homogeneous state to be unstable to inhomogeneous perturbations (*i.e.* in the presence of diffusion). With the notation  $\alpha = D_u/D_i$ , the mathematical conditions expressed by the above statement are [6]:

$$\begin{cases} \alpha F_x + G_y > 0, \\ (\alpha F_x + G_y)^2 - 4\alpha \det J_0 > 0, \end{cases} \quad (10)$$

where  $F_x \equiv \frac{\partial F}{\partial x}$  and  $G_y \equiv \frac{\partial G}{\partial y}$ , respectively. Eqs. (7) and (10) define the so-called Turing domain, *i.e.* the domain in the parameters' space in which the appearance of Turing structures is possible. Solving eqs. (10) we get:

$$\begin{cases} |R_d| > \frac{Z_0^2}{\alpha r}, \\ |R_d| > \frac{Z_0^2}{\alpha r} \left( \frac{2r\sqrt{\alpha}}{Z_0} - 1 \right). \end{cases} \quad (11)$$

From the first eq. (8) and the first eq. (11) a very well known result from the theory of Turing structures can be obtained:  $\alpha > 1$ . With other words, in any Turing system the activator is always diffusing slower than the inhibitor or substrate depleted (*i.e.* electrons diffuse faster than positive ions).

Eqs. (11) can be rewritten as follows:

$$|R_d| > \max \left\{ \frac{Z_0^2}{\alpha r}; \frac{Z_0^2}{\alpha r} \left( \frac{2r\sqrt{\alpha}}{Z_0} - 1 \right) \right\}. \quad (12)$$

Reuniting eqs. (9) and (12), the variation interval for the negative differential resistance of the ball of fire can be written as follows:

$$\max \left\{ \frac{Z_0^2}{\alpha r}; \frac{Z_0^2}{\alpha r} \left( \frac{2r\sqrt{\alpha}}{Z_0} - 1 \right) \right\} < |R_d| < \min \left\{ r; \frac{Z_0^2}{r} \right\}. \quad (13)$$

## V. CONCLUSIONS

Establishing the basic equivalent electric circuit for a plasma diode, at the anode of which a ball of fire is

formed, the reaction functions of the RD equations system can be easily derived. Applying the Turing formalism to that system of equations, it is shown that the ball of fire belongs to the same class of Turing structures like the Brusselator in chemistry. Moreover, one of the necessary conditions for the existence of the ball of fire at the anode surface in a plasma diode, resulting from experiments, is the negative sign of its differential resistance. At this conclusion also arrives the present approach, as a consequence of applying the Turing mathematical formalism. The variation interval for the negative differential

resistance of the ball of fire is also derived by imposing all the conditions defining the Turing domain in the parameters space.

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